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Molybdenum-99 Production via Fissile Solution Reactor and Electron Beam Accelerator

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INTRODUCTION

Currently, the U.S. supply of Molybdenum-99 (Mo-99) comes completely from foreign nations. No domestic source of Mo-99 has existed since the 1980s. The lack of a domestic Mo-99 supply is significant for many reasons. Mo-99 is an essential substance for the medical industry because it decays into technitium-99m (Tc-99m). Tc-99m is used in many medical imaging diagnostic tests for different types of cancer and heart conditions. However, Tc-99m only has a half-life of ~6 hours. Six hours is not enough time to transport and administer the isotope to patients undergoing this type of radioimaging. The solution to this complication is Mo-99. Mo-99 has a half-life of 66 hours which allows the material to be transported to the recipient, but does not allow for long term storage or stockpiling of the material. Therefore, Mo-99 must be "made-to-order". Not only is the lack of Mo-99 production a difficulty for the medical field, it is a national security risk. The dependency on other nations creates a difficult dynamic for the United States.

Many methods can be used to produce Mo-99, such as fission of ^{235}U, neutron capture of Mo-98, and various accelerator-based approaches [1, 2, 3]. Using an electron beam (E-beam) with a depleted or natural uranium (DU/NU) target will produce the (γ, n) reaction to interact with the fissile solution to produce Mo-99. For the purposes of this work a horizontal E-beam accelerator is designed for use with a uranyl nitrate, fissile solution reactor to understand the possible Mo-99 yields with different design parameters. Six case studies calculated with the Monte Carlo N-Particle code (MCNP) are explored and reported further in the following sections. Each case looks at the impact of changing one of six parameters, fissile solution type, solution base, solution concentration, uranium enrichment, and accelerator power and beam energy.

BACKGROUND

Production methods of Mo-99

Three main ways to create Mo-99 are neutron fission (n, f), neutron capture (n, γ), or accelerator based (γ, n) [3]. In an (n, f) reaction, a neutron fissions a ^{235}U atom that produces many fission products. One of which is Mo-99. Neutron capture involves shooting a neutron at Mo-98, and when the atom captures the neutron it becomes Mo-99. An accelerator-based approach can work in a variety of ways but still relies on a (γ, n) reaction. This study is concerned with the use of an accelerator produced E-beam to create the bremsstrahlung γ to induce a photofission reaction needed to create a fission reaction in the fissile solution. Similar designs to the design proposed here used a deuterium-tritium (DT) accelerator and a vertical fissile solution vessel [4]. During this process the system remains subcritical. By optimizing the Mo-99 yield conditions, multi-physics coupled modeling can be completed in the future to further understand the system characteristics in context of the nuclear-thermohydraulic feedback.

Two types of the fissile uranyl solution

The two fissile solutions investigated are uranyl nitrate and uranyl sulfate. To understand which is the best alternative among the two fissile solutions, one case uses uranyl sulfate to compare with the uranyl nitrate. Both options have pros and cons. Uranyl sulfate is relatively easy to prepare in comparison with uranyl nitrate. It also does not increase drastically in pH with irradiation due to radiolysis like uranyl nitrate will. Uranyl nitrate requires a nitric acid feed to prevent the fission products and uranium from precipitating out of the solution as the pH increases. However, a catalyst must be fed into the uranyl sulfate to prevent precipitation of uranyl peroxide and destroy the peroxide created [5, 6]. Each fissile solution is dissolved in a solution base, either H_2O or D_2O. Both base solutions are included in this work.

METHODOLOGY

Uranyl solution concentrations

To compare the different molar concentrations of uranyl solution the density of the uranyl nitrate solution has to be calculated as a function of molar concentration to be used in MCNP. Differing molar concentrations are used in each test case. An Oak Ridge National Laboratory report supplied the experimental density used in this study [7]. From this data the solution density was linearly extrapolated for solution concentrations up to the solubility limit. In Fig. 1 the data for the uranyl nitrate density based on solution concentration are shown.

MCNP modeling

The MCNP model geometry consists of a DU target, the fissile solution, a degassing region, and a stainless steel (SS316) vessel. A degassing region allows the radiolytic gases produced to flow elsewhere and eventually be removed from the system. The length and diameter of the horizontal cylindrical vessel are both 150 cm. Inside the vessel is a vacuum chamber that contains the DU target. It is 30 cm deep with a diameter of 32 cm. These dimensions can be manipulated to maximize the photonuclear reaction that occurs. A cylindrical target shape is used. It is an 8 cm long target with a 3 cm radius and a 4 cm cone shape cut out of the middle of the cylinder. In future studies, different target shapes will be modeled to optimize this aspect of the design. Fig. 2 illustrates the geometry of the horizontal fissile solution vessel and its corresponding materials.

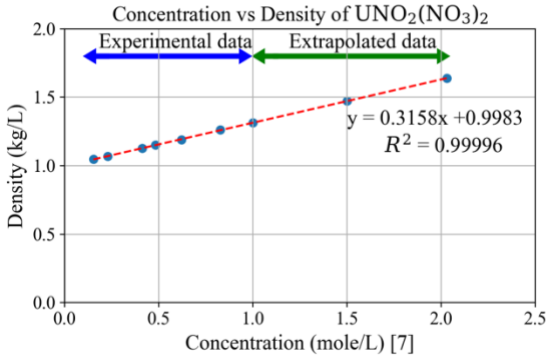

Fig. 1. Uranyl nitrate solution density vs molar concentration.

To find a starting point of the K_{eff} in the vessel a KCODE model was ran in MCNP to give a baseline criticality of each case. Each is subcritical as shown in the test matrix shown in table 1. The E-beam power and energy used are 40 MeV and 100 kW, which is equal to a beam intensity of 1.56E16 [e/s]. The low K_{eff} value (i.e. subcritical) is a key characteristic of pairing a generic fissile solution reactor with an accelerator. The reaction is easily controlled with the accelerator and is a very safe configuration. A drawback to this feature is that as the K_{eff} value is lowered. Therefore, so is the Mo-99 production yield. Finding the optimal relationship between these two features is an important future focus of this work.

Fission rates needed to calculate the Mo-99 yield are calculated with the F4 and F8 tallies. F4 tallies are used to find the average fission rate inside the specified cell. Fig. 3 shows the highest fission rates are seen near the DU target. The fission rate dissipates the further from the target the solutions is. These calculated fission rates directly translate to the activity calculation used to find the activity of Mo-99 present. It does not, however, account for the decay products produced during the reaction. The F8 tally does and will include the Mo-99 available from the decay of neighboring fission products as well as from fission. From this study an 8% increase in Mo-99 production yield with the F8 tally was observed when compared to the F4 tally. The isotope decay effects included in the F8 tally are the reason for the discrepancies of the two values, and are why both, F8 and F4, tallies are used.

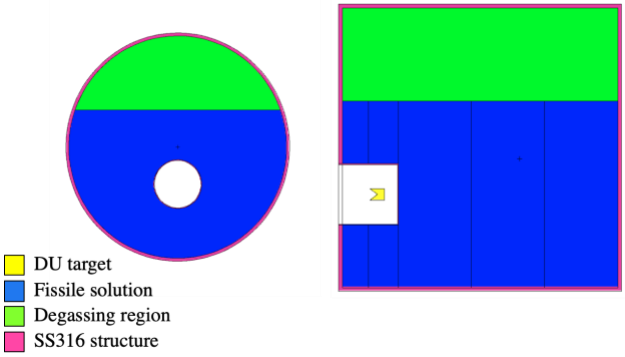

Fig. 2 MCNP geometry configuration.

Activity calculation

Mo-99 production yield is estimated based off of the activity of the whole fissile solution. Eq.(1) shows the equation used to calculate the yield in Curies.

$$Isotope\ produciton\ yield[t] = A_o\left(1 - exp^{-\lambda t}\right) \quad (1)$$

A_o is the starting activity of the isotope of interest, t is the time of the irradiation, and λ is the half-life of the isotope. The target irradiation time is 100 hours. Starting activity (A_o) was calculated using the fission rate and source particle intensity of the E-beam accelerator, both calculated in MCNP. This value is then multiplied by 6.01%, or the theoretical yield of Mo-99 from a (n, f) reaction. Essentially, these steps calculate the overall activity of the material and assumes 6.01% of it is from Mo-99.

Test matrix

As stated above, six test cases were performed to understand the effects of six design scenarios. In the test matrix below the conditions for each case are explicitly stated. If case 1 is the baseline, case 2 differs in its use of D_2O instead of H_2O as the base solution. Cases 3 and 4 are the same as case 1 except for the solution concentration. Case 1, 3, and 4 use a uranyl nitrate concentration of 400 g/L, 600 g/L, and 800 g/L, respectively. Case 5 differs from case 1 in the uranium enrichment used. Finally, case 6 uses uranyl sulfate as the fissile solution type instead of the uranyl nitrate. The conditions for the test matrix can be found in Table 1.

RESULTS

Fig. 4 shows the increase of Mo-99 production over the 100-hour time frame for each case. Case 5 is excluded

because the yield in Curies is a full order of magnitude higher than the other cases and will be addressed in the upcoming sections.

H_2O vs. D_2O

While comparing case 2 with all the other options it is clear case 2 produces much more Mo-99. The difference of this case to the others is D_2O, heavy water, was used as the solution base, opposed to the light water used in all the others. Heavy water has a lower absorption cross section than light water and allows for more interactions of neutrons with the fissile solution, and therefore leads to a higher fission rate.

Despite the higher fission rate other logistical problems of D_2O outweigh its benefits. Tritium is produced in high quantities with the irradiation of heavy water. A more complicated off-gas system would then be required. Due to safety and regulatory concerns, D_2O is considered to be a less preferable solution base to use for this application.

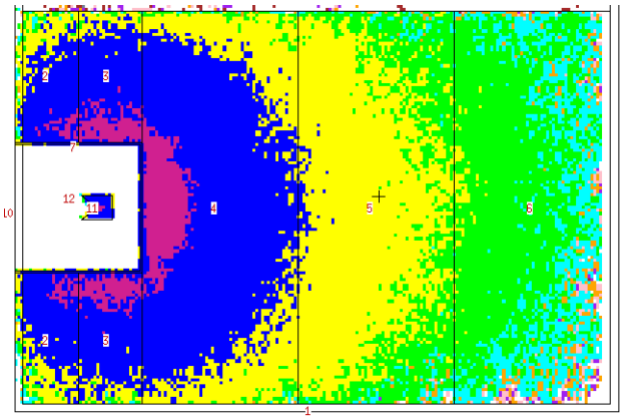

Fig. 3 Fission rate across vessel

U^{235} enrichment

Case 5 is the only case that used a 3wt% U-235/U enrichment over the natural uranium 0.7wt% U-235/U in the other samples. Fig. 5 shows the extent of the higher Mo-99 production with the higher enrichment. The final activity of Mo-99 in case 5 is 724 Ci, 12 times higher than the next highest value (case 2). Using 3wt% U-235/U enrichment may be slightly more difficult in handling due to the higher concentration of ^{235}U. However, it is still considered low enriched uranium, and is therefore a viable candidate for this application.

Uranyl nitrate vs. uranyl sulfate

Case 6 utilizes uranyl sulfate as the fissile solution in the model. From Fig. 4 it is clear that the lowest production values were in case 6. However, case 1 and case 6 were the only two with identical experimental conditions besides the uranyl type. While looking at these two cases the Mo-99 yield is not drastically different. Case 1 showed 30 Ci at 1.015 molar concentration and case 6 showed 18 Ci at 0.588 molar concentration. Hence it is expected that the production yield would be similar at the same molar concentration. Another consideration to be accounted for is the solubility limit in those two solution types. Per literature, nitrate-based solutions have relatively higher solubility limits over sulfate-based solutions. However, both cases produced the lowest amounts of Mo-99 out of all the options. It is clear the other parameters have a more impactful influence on the yield than uranyl types did.

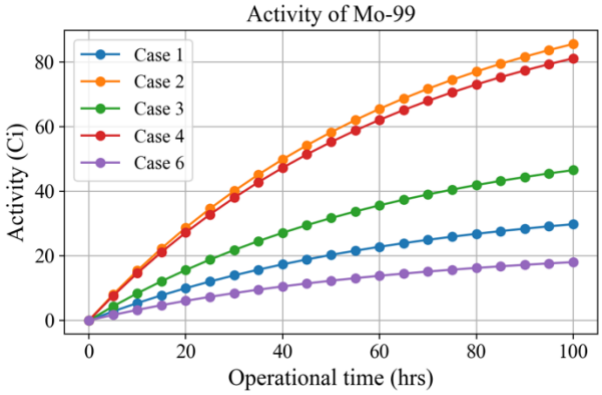

Fig. 4 Activity of Mo-99 in cases 1-4 and 6 over 100 hours.

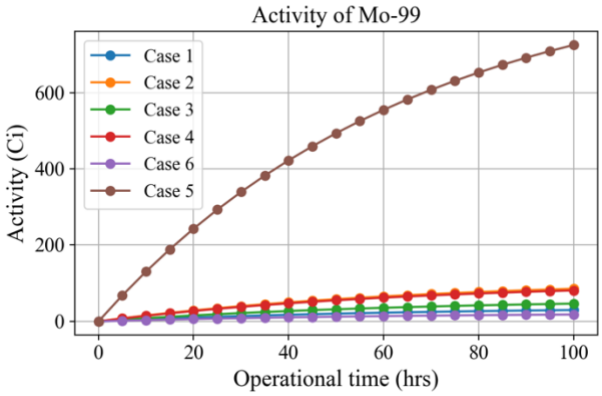

Fig. 5 Activity of Mo-99 in cases 1-6 over 100 hours.

Solution concentration

Of the three cases with different uranyl nitrate concentration, cases 1, 3, and 4, case 4 shows the most Mo-99 yield. The fissile solution in case 4 had a molar concentration of 2.031 M/L, or 800 g/L. The concentration and activity increase proportionately to one another. With the 30% increase in concentration between case 1 and case 3, there is a ~34% increase in activity. Similarly, with a 25% increase in case 3 to case 4 concentration, activity increases by ~23%. A higher uranyl nitrate concentration is desirable. However, the solubility limit of uranyl nitrate

TABLE 1 Test Matrix of six case studies and results

	Uranyl Type	U-235 Enrichment (wt%U_{235}/U)	Solution Concentration	Solution Base	Sol. Density [g/cc]	K_{eff}	Mo-99@100 hrs [Ci]	
							40 MeV, 100 kW	35 MeV, 10 kW
Case 1	$UO_2(NO_3)_2$	0.7 (Nat.)	1.015 M/L (400 g/Liter)	H_2O	1.31	0.1981	**30**	**3.4**
Case 2	$UO_2(NO_3)_2$	0.7 (Nat.)	1.015 M/L (400 g/Liter)	D_2O	1.31	0.4163	**85**	**9.7**
Case 3	$UO_2(NO_3)_2$	0.7 (Nat.)	1.523 M/L (600 g/Liter)	H_2O	1.47	0.2607	**46**	**5.2**
Case 4	$UO_2(NO_3)_2$	0.7 (Nat.)	2.031 M/L (800 g/Liter)	H_2O	1.64	0.3217	**60**	**6.8**
Case 5	$UO_2(NO_3)_2$	3 (Nat.)	2.031 M/L (800 g/Liter)	H_2O	1.64	0.8591	**724**	**82.5**
Case 6	$UO_2(SO_4)$	0.7 (Nat.)	0.588 M/L (215 g/Liter)	H_2O	1.185	0.12	**18**	**2.1**

in the base solution must be accounted for when the operating condition is determined. Precipitation of fission products is also a concern with this solution as the material becomes more acidic, so these traits must be optimized to find an ideal uranyl concentration in the fissile solution.

Accelerator beam characteristics

All six scenarios were modeled again using a smaller E-beam accelerator power, 35 MeV with 10 kW. The lower power translates to a beam intensity of 1.78E15 [e/s]. The final activity values are reported in Table 1. From this, it is obvious that the lower beam power produces less Mo-99. Beam intensity optimization is another aspect that will be studied in future work.

CONCLUSIONS

From the case studies used here the highest yield and most practical conditions are using a high concentration of uranyl nitrate, H_2O as a solution base, and a higher enrichment of ^{235}U if able. All of these should be combined with the higher E-beam energy conditions. The work presented here is part of the first steps towards creating a multi-physics coupled model where MCNP calculations will feed into computational fluid dynamics models. Future studies will look into the size, shape, and position of the DU target to see if further Mo-99 yield can be achieved.

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